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"Experimental and Modeling Studies of Interactions of Marine Aerosols and Clouds"

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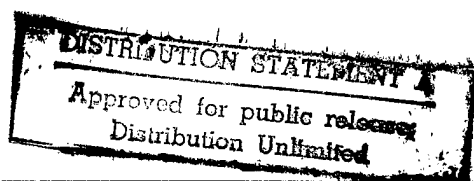
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**Experimental and Modeling Studies of Interactions
of Marine Aerosols and Clouds**

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Scientific Goals and Objectives of the Research

The primary goals of this research were to address the following key questions regarding marine aerosol / cloud interactions:

1. What factors control the abundance and vertical distribution of aerosol in the marine boundary layer?
2. How do these factors affect the formation and lifetime of marine clouds?

These questions have been addressed through a combination of modeling and experimental approaches, as described below.

The goal of the modeling component was to produce a model description of aerosol evolution and aerosol / cloud interaction that can be applied to a variety of marine boundary layer scenarios. The specific objective of this component was to develop a model for prediction of the effects of aerosol characteristics on cloud formation and evolution, and, in turn, the effects of cloud processes on the marine aerosol. The models built in this project can be used to study the role of aerosol in cloud modification, including ship tracks and indirect climate effects.

The goals of the experimental component were to develop new measurement techniques aimed at elucidating some important aerosol / cloud interactions, and to adapt and use these techniques in field work involving characterization of marine aerosol. A particular objective was the development of techniques for determination of size-dependent particle properties with high time resolution.

Approach / Tasks

Modeling Component. A main thrust of the modeling work has been the development of a new model concept, which can be used to examine processing of aerosol particles in marine stratocumulus clouds. Because of the large number of variables required to model simultaneous gas- and aqueous-phase chemistry, aerosol physics, and cloud microphysics, such coupled simulations are usually performed in zero-dimensional parcel models, driven by adiabatic updraft trajectories which pass once through the cloud. However, this approach cannot represent the cycling of parcels through a well-mixed stratocumulus-capped marine boundary layer, where in they experience both up- and downdrafts, and cycle in and out of cloud. Our

approach has been to perform large-eddy simulations of the marine boundary layer to obtain good simulations of parcel dynamics, then use trajectories diagnosed from those runs to drive a detailed parcel model. This unique approach can simulate the effects of cloud processing, both chemical and physical, on the aerosol population, which in turn feeds back into cloud evolution.

Experimental Component. Effort was focused on two fronts: (1) building our aerosol laboratory, establishing measurement techniques, and developing methodologies for calibration and testing of instruments; and (2) participation in field projects. As described in more detail below, key instrumentation was acquired through this grant, and deployed in an international marine aerosol field campaign, ACE-1. We also mounted our own campaign in the Rocky Mountains to field test our techniques. In the laboratory, we have focused on improving equipment that was available, customizing new instruments, and developing electron microscopy techniques that can be used to examine aerosol characteristics on a very detailed scale.

Results and Impact for Science

Modeling Component

1. Development of a coupled model of aerosol /cloud interactions

The use of the explicit-microphysics, large-eddy simulation (LES) version of the CSU Regional Atmospheric Modeling System (RAMS) to perform marine stratocumulus simulations is the state of the art in such modeling. This model can resolve small-scale turbulence features and produces realistic drop size spectra and drizzle (Kreidenweis et al., 1994). Under this grant, we have added a realistic aerosol treatment to this model (Richardson et al., 1995; Feingold and Kreidenweis, 1995; Kreidenweis and Feingold, 1995); this task was a challenge, particularly in the tracking of solute mass as it is transported through the drop spectrum and in the regeneration of altered aerosol from the evaporating drops. Our work defined some of the associated numerical difficulties, and identified approaches that can be used to minimize errors (Feingold et al., 1996).

2. Development of trajectory-driven parcel model and application to aerosol processing by marine stratocumulus clouds

In early work, we noted the impact of simplified treatments of boundary-layer dynamics on predicted aerosol concentrations (Kreidenweis, 1993; 1994), which led us to adopt the following approach. We employed the model described in (1) to examine the time scales for modification of the marine aerosol spectrum due to collision-coalescence processing alone (Kreidenweis et al., 1996; Feingold et al., 1996). This mechanism will deplete number concentration and increase the mean size of the cloud condensation nuclei (CCN), and is predicted to be quite effective at higher liquid water contents. We were able to examine a large parameter space in this study by implementing a new modeling concept which we developed: using diagnosed trajectories through the model cloud to drive an ensemble of parcels. Unlike simple adiabatic parcel models, this approach yields realistic estimates of total in-cloud residence time distributions

for the air parcels, which is important because the processes affecting the aerosol take place only in droplets.

In Feingold et al. (1996), we used simple arguments to compare the impact on the aerosol of collision-coalescence with that of aqueous-phase chemistry, and predicted conditions under which the two effects produced comparable changes in the CCN population. In continuing work, we have added gas- and aqueous-phase chemistry to the parcel model, and can now apply the trajectory approach to the coupled problem. We believe this modeling approach holds great promise for sensitivity studies of modification of clouds by anthropogenic perturbations (e.g., ship tracks; the so-called "indirect effect"), since it offers the high level of detail needed to simulate the aerosol / cloud interactions, yet does not sacrifice realistic boundary layer dynamics and thermodynamics.

3. Examination of the contribution of organic species to marine CCN

In Andrews et al. (1997), we performed a modeling study suggested by the observations of Novakov and Penner (1993), who found that organic species in marine aerosol samples at Puerto Rico contributed significantly to the number concentration of cloud condensation nuclei. This observation suggests that the emphasis that has been traditionally placed on sulfate species as CCN precursors may need to be broadened to include organics. However, the origin of the organic aerosol was not clear. The results of Andrews et al. (1997) suggested that the bulk of the organic contribution was likely to have been derived from local vegetation near the sampling site, skewing the interpretation of the aerosol as having strictly marine origins. These results have been used to redesign the sampling location to mitigate the effects of local vegetation. The role of organic species remains an open question, and we are continuing investigation into this issue.

Experimental Component

1. Development of aerosol laboratory

In the laboratory, our efforts have been largely focused upon the development of aerosol measurement capabilities. The acquisition of several important instruments through funding provided by ONR has played a key role. In particular, we purchased a differential mobility analyzer with the TSI scanning software, to enable us to make fast measurements of aerosol size distributions, and an ultrafine condensation nucleus counter, to enable us to detect particle nucleation events in the field. Under separate funding, we have purchased two aerosol generation systems that are used to calibrate our laboratory instruments.

Under ONR funding, we have also developed an aerosol volatility system that has been deployed in the field (Brechtel and Kreidenweis, 1994; 1996). The system is used to infer chemical composition of the aerosol, particularly to distinguish between ammoniated forms of sulfate. We have also built a hygroscopic tandem DMA system that can be used to determine the hygroscopic characteristics of particles of known size, although the system has not yet been deployed.

The CSU Dynamic Cloud Chamber is a unique facility that can be used to study aerosol / cloud interactions. Exploratory studies completed under separate funding, and further analyzed as part of this project (Jensen et al., 1995), have demonstrated the feasibility of using the chamber for the study of the initial effects of aerosol size and chemistry upon marine cloud formation. However, it was also shown that the chamber was not sufficiently well-controlled for low "ascent" rates characteristic of marine stratocumulus. Under this grant, effort has been devoted to the development of improved control of the CSU dynamic cloud chamber simulated adiabatic ascent. New control algorithms and new hardware have been developed and implemented, and have significantly improved the chamber performance. The completion of comprehensive documentation will facilitate the use of the chamber by new personnel.

2. Development of electron microscopy techniques

Graduate student Yalei Chen, assisted by Dr. Lynn McInnes of NOAA, developed techniques for the impaction of particles onto electron microscope grids and their subsequent examination and characterization via microscopy and X-ray analyses. These detailed techniques enable us to construct a three-dimensional particle size / composition distribution. This is of particular interest for marine CCN as it elucidates the role that sea salt plays in the *number* concentrations at small sizes, whereas traditional aerosol chemical collection techniques identify the contributions of chemical species to aerosol *mass*. In clean marine regions, sea salt is likely to be a major constituent of the CCN population, even in fine mode aerosol (McInnes et al., 1996). Another advantage of the electron microscopy technique is that samples can be obtained on very short time scales (~10 minutes), whereas impactor or filtration techniques require collection times of several hours or longer, decreasing the likelihood that the sample was obtained in a coherent air mass.

3. Rocky Mountain field experiment

Graduate student Fred Brechtel, assisted by Dr. Lynn McInnes and Dr. John Ogren of NOAA, designed and implemented a field experiment at Niwot Ridge, Colorado. Aerosol size distribution and ultrafine number concentrations were measured, to complement existing measurements of aerosol optical properties. Two objectives of the experiment were (1) to examine the source of ultrafine particles observed occasionally at Niwot Ridge, and (2) to field test the aerosol characterization system that was later deployed in ACE-1. We found that the ultrafine particles were most often associated with upslope conditions from the urban Colorado Front Range region (Brechtel et al., 1995), and heavily influenced by organic material. In this situation, volatility studies were too ambiguous to be used to identify constituents.

4. ACE-1 field experiment

In Jensen et al. (1996), we described results from our measurements in the ASTEX / MAGE experiment, which showed the response of the aerosol size distribution to changes in air mass source region. We expanded upon the suite of aerosol measurements in our contributions to the ACE-1 campaign, where our sampling was conducted at the remote marine location of Macquarie Island, midway between Tasmania and Antarctica. This remote site is ideal for examining the "background" marine aerosol, and indeed we found that even when the air mass

was continentally influenced, the heavy cloud cover and relatively long transport times to Macquarie Island modified the aerosol so that differences between "background" and "influenced" distributions were minimal (Brechtel et al., 1997).

A significant effort was devoted to the preparations for and deployment to the ACE-1 project. Graduate student Fred Brechtel traveled to Macquarie Island to make the measurements, and subsequent analyses have been carried out by him, Prof. Kreidenweis, and Dr. Lynn McInnes. We have found relationships between marine air mass source regions (NW, W, SW), dimethylsulfide concentrations, and the aerosol size distribution (Brechtel and Kreidenweis, 1996), which can be interpreted in terms of ocean sources of particles and the extent of processing of aerosol in marine clouds (Brechtel et al., 1997). Volatility studies indicate that sea salt occasionally accounted for most of the particle number above $0.07\text{ }\mu\text{m}$, which is the population active as CCN, although at other times sea salt dominated only in the particles larger than $0.2\text{ }\mu\text{m}$ (McInnes et al., 1996). The analysis of this rich data set is continuing; we are planning to submit a paper to the ACE-1 Special Issue of *J. Geophys. Res.* in mid-1997, and are presently working on several joint papers with other ACE-1 investigators.

Publications and Presentations Supported by this Award

- S.M. Kreidenweis, "A Simple Model of the Cloud-Topped Boundary Layer," presented at the Twelfth Annual Meeting of the American Association for Aerosol Research, Oak Brook, Illinois, 11-15 October, 1993.
- S. M. Kreidenweis, "A Simple Aerosol Model for Marine Boundary Layer Simulations," presented at the Annual AMS Meeting, Conference on Atmospheric Chemistry, Nashville, TN, January, 1994.
- F.J. Brechtel and S.M. Kreidenweis, "A Preliminary Study of the Volatility of Aerosol in the Colorado Rocky Mountains," presented at the Fourth International Aerosol Conference, Los Angeles, CA, August, 1994.
- S.M. Kreidenweis, W.A. Richardson, B. Stevens, and G. Feingold, "A Modeling Study of the Interaction Between Aerosol and Marine Stratus," presented at the Fourth International Aerosol Conference, Los Angeles, CA, August, 1994.
- T.L. Jensen, S.M. Kreidenweis, P.J. DeMott, D.C. Rogers, "Exploratory Dynamic Cloud Chamber Simulations of Initial Stratocumulus Formation," presented at the AMS Cloud Physics Conference, Dallas, TX, 15-20 January, 1995.
- W. Richardson, S.M. Kreidenweis, B. Stevens, and W. Cotton, "Aerosol processing in a two-dimensional hill top cloud using solute following microphysics: Eulerian and Lagrangian type models," presented at the Fourteenth Annual Meeting of the American Association for Aerosol Research, Pittsburgh, PA, 9-13 October, 1995.
- F.J. Brechtel, S.M. Kreidenweis, L.M. McInnes and J.A. Ogren, "Transport of anthropogenic aerosols and in-situ particle production at a remote mid-latitude mountain site," presented at the Fourteenth Annual Meeting of the American Association for Aerosol Research, Pittsburgh, PA, 9-13 October, 1995.
- G. Feingold and S.M. Kreidenweis, "Cloud processing of aerosol in stratocumulus: Collision-coalescence and regeneration," presented at the Fourteenth Annual Meeting of the American Association for Aerosol Research, Pittsburgh, PA, 9-13 October, 1995.
- S.M. Kreidenweis and G. Feingold, "Examination of aerosol cloud-processing potential in marine stratus clouds," presented at the Fourteenth Annual Meeting of the American Association for Aerosol Research, Pittsburgh, PA, 9-13 October, 1995.
- T.L. Jensen, S.M. Kreidenweis, Y. Kim, H. Sievering, A. Pszenny, "Aerosol Distributions in the North Atlantic Marine Boundary Layer During ASTEX / MAGE," *J. Geophys. Res.*, 101 (D2), 4455-4467, 1996.
- G. Feingold, S.M. Kreidenweis, B. Stevens, and W.R. Cotton, "Numerical simulations of stratocumulus processing of cloud condensation nuclei through collision-coalescence," *J. Geophys. Res.*, 101 (D16), 21,391-21,402, 1996.
- S.M. Kreidenweis, G. Feingold, B. Stevens, and W.R. Cotton, "Cloud processing of aerosol in the stratocumulus-capped marine boundary layer," *Proceedings of the 12th International Conference on Clouds and Precipitation, Zurich, Switzerland, 19-23 August, 1996.*

- F.J. Brechtel and S.M. Kreidenweis, "Observations of the aerosol number size distribution, total aerosol number concentration, and aerosol volatility at Macquarie Island during ACE-1," presented at the Annual Meeting of the American Association for Aerosol Research, Orlando, Florida, 14-18 October, 1996.
- L. McInnes, F.J. Brechtel and S.M. Kreidenweis, "Characterization of the non-volatile component of the marine aerosol during ACE-1," presented at the Annual Meeting of the American Association for Aerosol Research, Orlando, Florida, 14-18 October, 1996.
- F.J. Brechtel and S.M. Kreidenweis, "Observations of the aerosol size distribution and condensation nucleus concentration at Macquarie Island, Tasmania, during ACE-1," presented at the Fall Meeting of the American Geophysical Union, San Francisco, CA, December, 1996.
- F.J. Brechtel, S.M. Kreidenweis, L. McInnes, and H. Swan, "Aerosol characteristics at Macquarie Island, Tasmania, during ACE-1," Colorado State University, Department of Atmospheric Science Paper No. 633, 1997.
- E. Andrews, S.M. Kreidenweis, J.E. Penner, and S.M. Larson, "Potential origin of organic CCN observed at a marine site," *J. Geophys. Res.*, in review.